

Spin-current probe for phase transition in an insulator

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Spin fluctuation and transition have always been one of central topics of magnetism and condensed matter science. Experimentally, the spin fluctuation is found transcribed onto scattering intensity in the neutron scattering process, which is represented by dynamical magnetic susceptibility and maximized at phase transitions. Importantly, a neutron carries spin without electric charge, and it can bring spin into a sample without being disturbed by electric energy, although large facilities such as a nuclear reactor is necessary. Here we show that spin pumping, frequently used in nanoscale spintronic devices, provides a desktop micro probe for spin transition; spin current is a flux of spin without an electric charge and its transport reflects spin excitation. We demonstrate detection of antiferromagnetic transition in ultra-thin CoO films via frequency dependent spin-current transmission measurements, which provides a versatile probe for phase transition in an electric manner in minute devices.

A spin current refers to a flow of spin angular momentum of electrons in condensed matter¹. There are types of spin currents, including a spin current carried by conduction electrons and one carried by spin waves¹. The former type of spin current, conduction-electron spin current, can be detected by using the inverse spin Hall effect (ISHE)²⁻⁷, the conversion of a spin current into electric voltage via the spin-orbit interaction in a conductor, typically in Pt. Although conduction-electron spin current can reside only in metals and semiconductors, the latter type of spin current, called spin-wave spin current, can exist even in insulators^{4,8}. In fact, spin-wave spin currents have been studied in magnetic insulators and, very recently, in antiferromagnetic alloys and insulators⁹⁻¹⁵.

For spin-current generation, one of the most versatile and powerful methods is spin pumping, an induction of spin current from magnetization precession in a magnet into an attached metal via the exchange interaction at the interface^{4–6, 16–22}. Spin pumping was found to drive spin current also from magnetic insulators, such as $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ^{4, 6, 16–22}.

Various powerful theories have been constructed to describe the spin pumping phenomena^{23–25}. They commonly predict that the efficiency of spin pumping is sensitive to dynamical magnetic susceptibility at the interface between a magnet and a metal in a spin pumping system. Therefore, spin pumping is sensitive to interface magnetic susceptibility, while standard magnetometry probes bulk properties which often hides interface signals. This raises an interesting hypothesis: when a very thin sample film is inserted at the interface of a spin pumping system, spin pumping may reflect the dynamical susceptibility, which is directly related to spin fluctuation according to the fluctuation-dissipation theory, of the inserted thin sample film. Here we show that this is the case by using an antiferromagnetic transition in an ultra-thin film of CoO, and that spin pumping becomes an *in-situ* micro probe for magnetic phase transition.

Results

Sample description. Figure 1c is a schematic illustration of the sample system used in the present study; we inserted an antiferromagnetic CoO thin film at the interface between $\text{Y}_3\text{Fe}_5\text{O}_{12}$ and Pt layers in a typical spin-pumping system $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$ to form $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$. At low temperatures, CoO exhibits antiferromagnetic order²⁶. Here, $\text{Y}_3\text{Fe}_5\text{O}_{12}$ is a typical spin pumping material,

in which spin current is emitted from $\text{Y}_3\text{Fe}_5\text{O}_{12}$ when magnetization precession in $\text{Y}_3\text{Fe}_5\text{O}_{12}$ is excited^{4,6}. Pt is used as a spin-current detector based on ISHE, in which a spin current is converted into an electric voltage in the Pt perpendicular to the spin-current spin polarization direction^{2-4,6}. When magnetization precession in $\text{Y}_3\text{Fe}_5\text{O}_{12}$ is excited by a microwave application, spin pumping is driven and then a spin current is injected from the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ layer into the Pt layer across the thin CoO layer²³⁻²⁵.

Spin pumping signal of $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ system. Figure 2a shows the magnetic field dependence of microwave absorption spectra of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ at various temperatures measured when a 5 GHz microwave is applied. At $T=300$ K, absorption peaks appear around $H_{\text{FMR}} = \pm 1.2$ KOe, which correspond to ferromagnetic resonance (FMR) in the $\text{Y}_3\text{Fe}_5\text{O}_{12}$. With decreasing temperature, H_{FMR} is observed to slightly decrease, which is due to the temperature dependence of magnetization in the $\text{Y}_3\text{Fe}_5\text{O}_{12}$. The microwave absorption power P_{ab} at H_{FMR} is almost constant with changing T .

In Fig. 2b, we show the voltage V generated in the Pt layer in a simple $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$ spin-pumping system without a CoO layer measured with applying a 5 GHz microwave. At the FMR field H_{FMR} , a clear voltage peak appears at all temperatures. The sign of the peak voltage V_{ISHE} is reversed by reversing the polarity of the applied magnetic field, showing that the voltage peak is due to ISHE induced by spin current pumped from the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ layer^{2-4,23-25}.

Figure 2d shows the temperature dependence of the peak voltage V_{ISHE} for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$ film without a CoO layer. V_{ISHE} decreases monotonically with decreasing the temperature. This

monotonic decrease is attributed to the decrease in the resistivity of Pt and the increase in the magnetization damping in $\text{Y}_3\text{Fe}_5\text{O}_{12}$.

Nevertheless, by inserting a CoO layer in the simple spin-pumping system, a clear unconventional peak structure appears in the temperature dependence of the ISHE signal (Fig. **2c**). Figure **2e** shows the temperature dependence of the voltage peak intensity V_{ISHE} for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer film. The V_{ISHE} for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer film exhibits a clear peak at $T = 200$ K, which is quite different from that for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$ bilayer film (Fig. **2d**). The V_{ISHE} peak temperature is comparable to the Néel temperature of the CoO layer determined by an X-ray magnetic linear dichroism (XMLD) measurement using a synchrotron facility as shown in Fig. **2f** (detailed data please see Supplementary Note 1, Supplementary Figure 1 and Supplementary Figure 2). Furthermore, the temperature dependence of V_{ISHE} is similar to that of the magnetic susceptibility in a bulk CoO, in which the susceptibility is maximized around the Néel temperature $T_N^{27,28}$.

Spin pumping with different CoO layer thickness. Figure **3a** shows the temperature dependence of V_{ISHE} measured for various thicknesses of the CoO layer in $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer films. In CoO films, the Néel temperature is known to decrease with decreasing the thickness of the film due to the finite size effect^{29,30}. The observed CoO-thickness dependence of the peak temperature is consistent with this feature: the peak temperature decreases with decreasing the CoO layer thickness. All the results show that the V_{ISHE} peak position indicates the Néel temperature of the CoO layer, and that the V_{ISHE} enhancement around the antiferromagnetic transition can be related to the CoO-film susceptibility enhancement, which is a good measure for spin fluctuations.

Spin pumping signal of $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{NiO}/\text{Pt}$ system. To check the universality of the phenomenon, we measured another antiferromagnet: a NiO film (1.5 nm) in a $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{NiO}/\text{Pt}$ system and found a similar peak structure and temperature dependence of V_{ISHE} (Fig. **3b**). The peak position is consistent with the previous study on the Néel temperature in ultra-thin NiO films³¹. Furthermore, when a Cu layer is inserted between the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ and the CoO (NiO) layers, we observed similar peak structures in the T -dependent ISHE signal (Supplementary Note 2 and Supplementary Figure 3). This result suggests that the direct exchange coupling between $\text{Y}_3\text{Fe}_5\text{O}_{12}$ and an antiferromagnet is not necessary for the V_{ISHE} enhancement around Néel temperatures. Although observing such a phase transition in a single ultra-thin film was impossible without using large synchrotron facilities and a special XMLD spectrometer^{32–34}, our present method provides a way to probe it by a table-top experiment.

Discussion

Our present study stimulates further investigations of not only the spin transport near magnetic phase transitions but also microscopic spin-transport properties in antiferromagnetic systems. In antiferromagnetic insulators, incoherent thermal magnons and coherent Néel order parameter dynamics¹² are considered to be responsible for transporting spins. Our experimental results in Fig. **3**, however, show that V_{ISHE} is strongly suppressed toward lower temperatures in both cases of $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ and $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{NiO}/\text{Pt}$ systems; V_{ISHE} at 10 K is much less than that at T_{N} . Also, we notice that this feature was confirmed by some recent studies^{35,36}. These results indicate that the spins are transported dominantly by incoherent thermal magnons rather than coherent

Néel dynamics. At high temperatures, thermal magnons continuously evolve into thermal spin fluctuations, which would transport spin current above Néel temperature.

Such thermal spin dynamics both below and above T_N are well described by a bosonic auxiliary particle method³⁷. Using this method, the spin conductivity in an antiferromagnetic insulator was shown to be maximized near its Neel temperature³⁸ exactly like our V_{ISHE} (Fig. **2e** inset). Since V_{ISHE} measures spin moments transferred across magnetic insulators, its enhancement directly reflects that of the spin conductivity. The spin conductivity and the magnetic susceptibility are in principle different quantities. However, their temperature dependences are rather similar because both are dominated by spin excitations with zero momentum transfer. Therefore, V_{ISHE} in our experimental setup is a good measure for the spin dynamics and transition.

Moreover, another significant remark on our result which requires further theoretical understanding is the frequency dependence of the spin-pumping behavior. Figure **4a** shows the microwave frequency f as well as the temperature T dependence of $V_{\text{ISHE}}/P_{\text{ab}}$ for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer film, where V_{ISHE} is normalized by the FMR microwave absorption P_{ab} . For all the frequencies, $V_{\text{ISHE}}/P_{\text{ab}}$ shows peaks around the Néel temperature $T_N \sim 130$ K. As seen in Fig. **4c**, $V_{\text{ISHE}}/P_{\text{ab}}$ exhibits different f dependence at different temperatures, namely $V_{\text{ISHE}}/P_{\text{ab}}$ strongly depends on f only near $T_N \sim 130$ K, but it weakly depends on f at temperatures far from T_N . Such a strong frequency dependence implies that the observed phenomena reflect dynamical properties. Similar frequency dependence was also observed in the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{NiO}/\text{Pt}$ trilayer film, but it was absent in the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$ bilayer film without antiferromagnetic layers (Supplementary Note 3

and Supplementary Figure 4), showing that the observed prominent frequency dependence is characteristic to the antiferromagnetic layers near T_N . This type of strong frequency dependence cannot be explained by the coherent Néel dynamics or simple thermal magnons alone. First of all, coherent precession of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ can pump antiferromagnetic magnons at the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ /antiferromagnetic interface, in analogy to the conventional spin pumping at the ferromagnet/metal interfaces¹². Secondly, the FMR dynamics inside of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ itself should pump thermal magnons, which can subsequently diffuse across the structure, inducing the ISHE signal. This latter process relies on the breaking of the $\text{SU}(2)$ symmetry of magnetic dynamics³⁹, which would be progressively enhanced with the increased ellipticity of the coherent dynamics in $\text{Y}_3\text{Fe}_5\text{O}_{12}$. Increased ellipticity of the Kittel mode in $\text{Y}_3\text{Fe}_5\text{O}_{12}$ at lower frequencies can thus be responsible for the enhancement of the conversion of the coherent precession into thermal magnons, leading to the observed increase in the signal. Alternatively, the finite life time of thermal magnons could be a source of the observed frequency dependence as in the case of the frequency dependent spin conductance³⁸. In principle, the magnon life time depends on temperature and other intrinsic and extrinsic effects. A detailed theoretical analysis of the interplay of the coherent and incoherent magnetic dynamics in our heterostructure is, however, beyond the scope of this work. Constructing a comprehensive theory for spin-current transport in magnetic heterostructures is an important outstanding task for the development of novel spintronics based on quantum magnets.

Methods

Preparation of $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ samples. A 3 μm -thick single-crystalline $\text{Y}_3\text{Fe}_5\text{O}_{12}$ film was grown on a (111) $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ wafer by a liquid phase epitaxy method at 1203 K in a $\text{PbO}-\text{B}_2\text{O}_3$ based flux. All the samples were cut from a same wafer into $1.5 \times 3 \text{ mm}^2$ in size. CoO films with different thicknesses were coated on the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ film by an rf magnetron sputtering method. All the CoO films were prepared at 1073 K to restrain the formation of cobalt oxide with other valence states and to improve the crystallinity. Then, 10 nm-thick Pt films were put on the top of the CoO films with a Hall-bar structure by an rf magnetron sputtering method.

Sample characterization. Crystallographic characterization for samples was carried out by a X-ray diffractometry and transmission electron microscopy (TEM). A TEM image shows that the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ film is of a single-crystal structure, and CoO and Pt layers are nearly epitaxially grown on the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ film (Fig. 1d). An X-ray photoelectron spectroscopy (XPS) method was used to confirm the chemical valence of the CoO layer (Fig. 1e).

Spin-pump experimental set-up. The spin-pumping measurement was performed in a PPMS system, Quantum design inc.. To excite FMR in the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ layer, microwave was applied by using a coplanar waveguide. The voltage signal between the ends of the Pt layer was measured by using a lock-in amplifier. Temperature dependence measurement was carried out from 10 K to 300 K, after cooling samples from room temperature to 10 K in a 5000 Oe magnetic field.

Data availability. All relevant data are available from the corresponding author on request.

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Author contributions

Z.Q. and D.H. designed the experiment, fabricated the samples, collected all of the data and analyzed the data. E.S. supervised this study. J.L., Z.Q.Q., E.A., A.T.N. and A.T. performed the

XMLD and XMCD measurements. K.S., S.O. and Y.T. contribute theoretical suggestions. K.U. helped the low temperature experiment. Z.Q., D.H., E.S. S.O. K.S., K.U. and Y.T. wrote the manuscript. All the authors discussed the results and commented on the manuscript.

Figure 1 Concept and sample set-up. **a**, Inelastic scattering of polarized neutrons through an antiferromagnetic system. **b**, Spin-current transmission through an antiferromagnetic system. **c**, Experimental set-up of the spin pumping measurement for the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer device. J_s denotes spin current injected from the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ layer into the Pt layer through the CoO layer by spin pumping, which is detected as a voltage signal via the inverse spin Hall effect in the Pt layer. **d**, A crosssectional TEM image of a $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer device. The scale bars of the image is 10 nm. **e**, A Co 2p XPS spectrum and Gaussian fitting analysis for the CoO layer in the $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{CoO}/\text{Pt}$ trilayer device.

Figure 2 Spin pumping detection of antiferromagnetic transition. **a**, Magnetic field (H) dependence of microwave absorption power (P) for a $\text{Y}_3\text{Fe}_5\text{O}_{12}$ film (3 μm in thickness) at various temperatures. P_{ab} denotes absorption power at ferromagnetic resonance (FMR) field. **b**, Magnetic field (H) dependence of electric voltage (V) generated in the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (3 μm)/Pt (10 nm) bilayer film at various temperatures. **c**, Magnetic field (H) dependence of electric voltage (V) generated in the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (3 μm)/CoO (6 nm)/Pt (10 nm) trilayer film at various temperatures. V_{ISHE} denotes the voltage signal at the FMR field. **d**, Temperature dependence of V_{ISHE} for the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (6 μm)/Pt (10 nm) bilayer film. **e**, Temperature dependence of V_{ISHE} for the $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (3 μm)/CoO (6 nm)/Pt (10 nm) trilayer film. The inset shows the theoretical prediction of the spin conductance versus temperature in an antiferromagnetic system with $S=1/2$ ³⁸. Coefficient D denotes the spin

conductance at a given frequency scaled by its maximum value D_{\max} . **f**, Temperature dependence of the XMLD signal ΔR_{L3} for the $Y_3Fe_5O_{12}$ (3 μm)/CoO (6 nm)/Pt (1 nm) trilayer film (details are shown in supplementary Note 1). The error bars in **d**, **e** and **f** represent the s.d. of multiple measurements at the same condition.

Figure 3 Temperature dependence of spin pumping signals in different systems.

a, Temperature dependence of V_{ISHE} for $Y_3Fe_5O_{12}$ /CoO/Pt trilayer films with different CoO-layer thicknesses (d_{CoO} =3, 6 and 10 nm). **b**, Temperature dependence of V_{ISHE} for a $Y_3Fe_5O_{12}$ /NiO(1.5 nm)/Pt film. The dash lines in **a** and **b** denote the peak positions.

Figure 4 Frequency dependence of spin pumping signals in $Y_3Fe_5O_{12}$ /CoO/Pt. **a**,

A pseudo-colour plot of V_{ISHE}/P_{ab} as a function of the temperature T and the microwave frequency f for the $Y_3Fe_5O_{12}$ (3 μm)/CoO (3 nm)/Pt (10 nm) trilayer film. **b**, Temperature (T) dependence of V_{ISHE}/P_{ab} at various exciting microwave frequencies. **c**, Exciting microwave frequency (f) dependence of V_{ISHE}/P_{ab} at various temperatures. We did not show the data for frequencies lower than 4 GHz because the magnetization precession can be modulated due to the three-magnon interaction when $f < 4GHz^{40}$.